Relationships among tracer ages

Darryn W. Waugh
Department of Earth and Planetary Sciences, Johns Hopkins University, Baltimore, Maryland, USA

Timothy M. Hall
NASA Goddard Institute for Space Studies, New York, New York, USA

Thomas W. N. Haine
Department of Earth and Planetary Sciences, Johns Hopkins University, Baltimore, Maryland, USA

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[1] Measurements of chemical tracers whose spatial gradients are primarily due to the time dependence of sources and/or sinks are often used to define "tracer ages" in an effort to diagnose transport. However, a major problem with interpreting these tracer ages is that different tracers can yield different ages, and at present, it is not clear what aspects of the transport are measured by the different tracers. We use the concept of a distribution of transit times to compare the timescales derived from different tracers, including CFCs, tritium-helium, and radioactive tracers. By performing a systematic study over a range of transit time distributions we examine under what conditions two tracers yield similar or different ages. It is shown that there can be significant differences in tracer ages and that in general, tracer ages are not fundamental timescales of the flow. Furthermore, even if ages from two tracers are similar these ages can be very different from the mean (ideal) age or the age of a third tracer. It is also shown that significant temporal variations in tracer ages can occur for steady transport and that these changes are of similar magnitude to the changes in CFC and tritium-helium ages observed in the North Atlantic and North Pacific over the 1980s and 1990s. Accounting for the changes in tracer ages caused by steady transport is necessary before attributing changes in tracer ages to changes in transport. The possibility of using the differences in ages from different tracers to infer information about the transit time distribution is also examined. It is shown that two tracer ages can constrain the first two moments (mean age and width) of the distribution, but how tightly these are constrained depends on the tracers used, the certainty of the age calculations, and the flow characteristics. *INDEX TERMS:* 4808 Oceanography: Biological and Chemical: Chemical tracers; 4283 Oceanography: General: Water masses; 1832 Hydrology: Groundwater transport; *KEYWORDS:* ocean tracers, ocean transport, tracer ages, transit time distributions

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1. Introduction

[2] The timescales for transport from a boundary to the interior of a fluid system are fundamental descriptions of the flow. Chemical tracers with known sources and sinks can be useful in estimating these timescales. To this end, tracers have been used to derive "ages" in a wide range of geophysical settings, for example, oceans [e.g., *Fine*, 1995; *Schlosser and Smethie*, 1996; *Schlosser et al.*, 2001], lakes [e.g., *Weiss et al.*, 1991], groundwater [e.g., *Plummer et al.*, 1993; *Cook and Solomon*, 1997], and the stratosphere [e.g., *Waugh and Hall*, 2002]. These "ages" are commonly assumed equivalent, or closely related to, the underlying flow timescales. They have been used therefore to diagnose flows, as well as to summarize the infiltration of pollutants and test transport in models. A major drawback lending ambiguity to the interpretation is that "age", as typically defined, varies with tracer. In many applications the tracer-independent features of the underlying flow diagnosed by age have not been clearly separated from tracer-specific properties.

[3] As a result of mixing, there is no single timescale, or "transit time", for surface water to a given interior location. Instead, there is a distribution of transit times. Each tracer, because of its different boundary condition or decay rate, weighs features of the distribution differently and so in general different tracers yield different ages. It is the distribution, rather than the particular tracer age, that is a fundamental descriptor of the flow. The fact that different tracer ages can differ is well known, but the possible range of differences and dependence of the differences on the transport characteristics are not well quantified.

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[4] Here we use the concept of a transit time distribution to perform a systematic examination of the relationship between ages derived from different tracers. We consider a wide range of tracers, including conserved tracers with time varying surface concentrations (e.g., chlorofluorocarbons (CFCs)) and tracers with radioactive decay (e.g., tritium, argon, and radiocarbon), and transit time distributions that represent a wide (though necessarily incomplete) range of flows. Several other studies have looked at age-age relationships in observations and simple advection-diffusion models, for example, *Thiele and Sarmiento* [1990], *Doney et al.* [1997], *Ekwurzel et al.* [1994], and *Sonnerup* [2001]. Also, *Wunsch* [2002] has recently examined tracer ages and transit time distribution in some simple, analytical and numerical, models.

[5] The existence of a distribution of transit times results not only in differences between ages from different tracers but can also result in temporal changes in tracer ages for steady transport. The possibility that steady transport can produce the observed change in the tritium-helium age in the North Atlantic subtropical gyre has already been discussed by *Robbins and Jenkins* [1998]. Here, we use the transit time distribution framework to examine the temporal change in tracer ages. Again we consider a range of transit time distributions, and examine the sensitivity of the temporal evolution to the characteristics of the transit time distribution.

[6] Although the fact that different tracers yield different ages makes interpretation of these ages difficult, it does raise the possibility that ages from multiple tracers may provide information on the distribution of transit times, as has been noted previously, for example, by *Cook and Bohlke* [1999]. Using the above sensitivity studies we explore whether various pairs of tracer ages can be can be utilized to infer information about the shape of the transit time distribution. Some closely related analysis was done by Alfred Putzka and appears in an unpublished manuscript "Age distributions and transient tracer dating of ocean waters" (hereinafter referred to here as A. Putzka (unpublished manuscript, 1999)). Also, *Klatt et al.* [2002] used a time series of CFC measurements in the Weddell Sea to infer aspects of the transit time distribution.

[7] The various quantities, including the transit time distribution and the mean age, used to define the transport timescales are defined in the next section, as are the various "ages" calculated from tracers. In section 3 we examine the tracer ages for several families of idealized tracers in which there are simple analytical expression for their growth or decay, and we can derive approximate equations for the tracer ages. Realistic tracers (e.g., CFCs, tritium, and argon 39) are then considered in section 4, where we examine the relationships between the ages derived from these tracers for a wide range of transit time distributions. The temporal variation of tracer ages for steady transport is then examined in section 5, and compared with several recent observational studies. In section 6 we examine the possibility of constraining the transit time distribution, in particular the first two moments, using multiple tracer ages. The sensitivity of the analysis in the previous sections to the assumed form of the transit time distribution is examined in section 7, and concluding remarks are in section 8.

2. Tracers and Age

2.1. Transit Time Distributions

[8] As discussed in the Introduction, there is a distribution of a transit times for transport from the surface to a given interior location within a fluid system. The existence of this distribution has been discussed in the context of transport in the oceans, groundwater, and the atmosphere [e.g., *Beining and Roether*, 1996; *Delhez et al.*, 1999; *Deleersnijder et al.*, 2001; *Khatiwala et al.*, 2001; *Haine and Hall*, 2002; *Cook and Bohlke*, 1999; *Waugh and Hall*, 2002]. The distribution has been referred to by various names, including transit time distribution, age frequency distribution, age distribution, and age spectrum. Here, we shall refer to it as the "transit time distribution", or "TTD" for short.

[9] Mathematically the TTD is a type of Green function that propagates the tracer mixing ratio at the surface $c_0(t)$ into the interior [*Hall and Plumb*, 1994]. The concentration time series at interior location r with transit time distribution $\mathcal{G}(r, t)$ is given by

$$c(r,t) = \int_0^\infty c_0(t-t')\mathrm{e}^{-\lambda t'}\mathcal{G}(r,t')dt', \tag{1}$$

where the $\mathrm{e}^{-\lambda t'}$ term is for the radioactive decay of tracers with decay rate λ. $\mathcal{G}(r, t)dt$ is the mass fraction of the fluid parcel at r that was last at the surface a time t to $t + dt$ ago. Note that we have assumed steady state conditions and spatially uniform surface conditions, but time-dependent flows and nonuniform surface conditions can be accommodated [e.g., *Holzer and Hall*, 2000; *Haine and Hall*, 2002]. Given $\mathcal{G}(r, t)$, together with $c_0(t)$ and λ, the concentration time series $c(r, t)$ can be determined from equation (1). Hence the TTD concept provides a framework to compare the ages derived from tracers with different surface time series and decay rates.

[10] In the analysis below it is helpful to consider the temporal moments of the TTD. The nth moment is

$$m_n = \int_0^\infty \xi^n \mathcal{G}(\xi)d\xi. \tag{2}$$

By definition, the zeroth-order moment of $\mathcal{G}$ equals unity for a closed domain. The first moment

$$\Gamma \equiv m_1 = \int_0^\infty \xi\mathcal{G}(\xi)d\xi, \tag{3}$$

is known as the "mean age". For higher moments it proves to be more useful to consider moments about Γ (centered moments) rather than about the origin. The nth centered moment is

$$\mu_n = \int_0^\infty (\xi-\Gamma)^n \mathcal{G}(\xi)d\xi. \tag{4}$$

By definition, the first centered moment vanishes, but the second moment μ_2 is generally nonzero and provides a measure of the spread of the TTD. Following *Hall and Plumb* [1994], we define the "width" Δ as

$$\Delta^2 \equiv \frac{1}{2}\mu_2 = \frac{1}{2}\int_0^\infty (\xi-\Gamma)^2 G(\xi)d\xi. \tag{5}$$

Note that there are simple relationships between the moments about the origin and those about the mean, for example, $\mu_2 = m_2 - m_1^2$.

2.2. Tracer Ages

[11] Timescales (“ages”) can be derived from several different types of tracers. The calculation of these ages varies between the different types of tracers, and we briefly outline these calculations below. Again different terminology has been used in the literature for the timescale derived from tracers, including “apparent age”, “ventilation age”, “tracer time lag”, and “tracer age”; we shall in general use tracer age.

[12] One class of tracers are conserved tracers with monotonically increasing (or decreasing) surface concentrations. Included in this class are the chlorofluorocarbons (e.g., CFC-11, CFC-12, and CFC-113) and carbon tetrachloride (CCl_4), all of which were increasing until (at least) the early 1990s, see Figure 1a. From these tracers a “concentration” tracer age can be defined as the elapsed time since the surface concentration was equal to the interior concentration. Mathematically, this age τ_{conc} is given by

$$c(t) = c_0(t - \tau_{conc}), \tag{6}$$

where $c(t)$ is the interior and $c_0(t)$ the surface concentration.

[13] A related age can be defined if the ratio of two conserved tracers is monotonically increasing (or decreasing), for example, CFC ratios shown in Figure 1b. This “ratio” tracer age is the elapsed time since the ratio $R(t)$ of two tracer concentrations at the surface was equal to the interior ratio:

$$R(t) = R_0(t - \tau_{ratio}). \tag{7}$$

The ratio age has the advantage that it is unaltered by mixing with tracer-free water.

[14] Another class of tracers are those undergoing radioactive decay, for example, tritium, argon, natural radiocarbon. If the surface concentration is approximately constant in space and time then a “radioactive” tracer age can be defined as

$$\tau_{radio} = \lambda^{-1} \ln\left(\frac{c_0}{c}\right), \tag{8}$$

where c_0 is the fixed surface concentration, c the interior concentration, and λ the radioactive decay constant ($\lambda = \ln(2)/T$, where T is the half-life of the tracer). There are significant temporal variations in the surface concentration of tritium because of atmospheric bomb testing in the mid-1960s, and a modified form of equation (8) has to be used to define an age from tritium [e.g., *Jenkins and Clarke*, 1976]

$$\tau_{\mathrm{He}} = \lambda^{-1} \ln\left(\frac{{}^3\mathrm{H} + {}^3\mathrm{He}^{\dagger}}{{}^3\mathrm{H}}\right) \tag{9}$$

where ${}^3\mathrm{He}^{\dagger}$ (“excess” or tritiogenic ${}^3\mathrm{He}$) is the component of ${}^3\mathrm{He}$ that has come from ${}^3\mathrm{H}$ decay, and $\lambda = 0.05576$ years^{-1} is the decay constant of ${}^3\mathrm{H}$ (half-life $T = 12.43$ years) [*Unterweger et al.*, 1980].

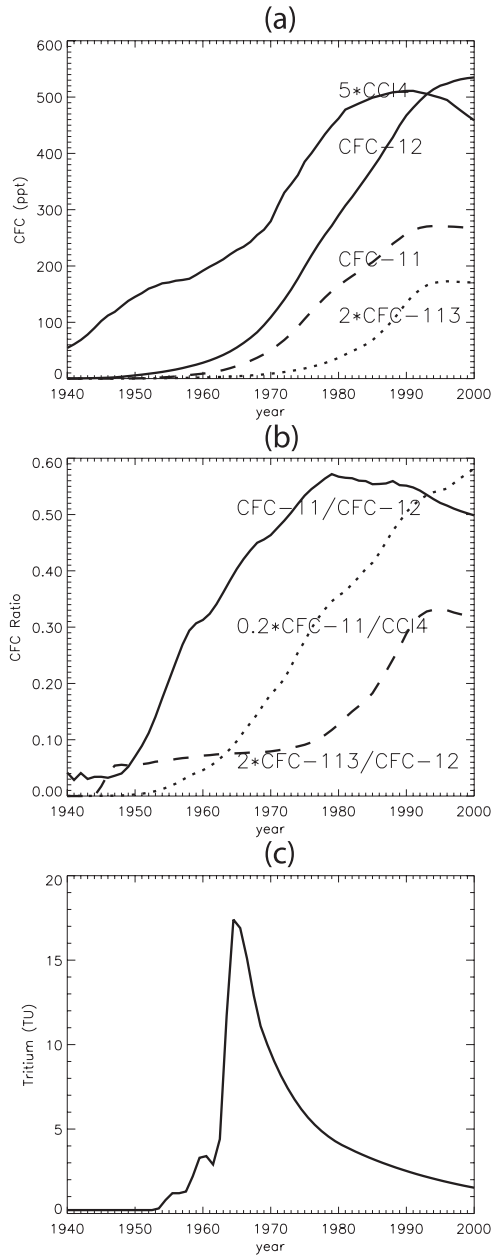


Figure 1. Time series of (a) atmospheric concentrations of halocarbons (CFC-11, CFC-12, CFC-113, and CCl_4), (b) ratios of halocarbons (CFC-11/CFC-12, CFC-113/CFC-12, and CFC-11/CCl_4), and (c) surface water tritium.

[15] If the circulation is purely advective then all the tracer ages defined above are identical, and equal to the time for advection from surface to the interior location. However, because of mixing the above ages are not generally the same. Below, we use the TTD concept to provide some insight into the differences between the different tracer ages.

3. Idealized Tracers

[16] Before considering realistic tracers we consider idealized tracers for which there are simple analytical expressions for the temporal variations in the surface con-

centration, and approximate equations for the tracer age in terms of the moments of the TTD.

[17] We first consider conserved tracers with exponential growth λ, i.e., $c_0(t) = e^{\lambda t}$. Substituting this into equation (1) and combining with equation (6) we have

$$\tau = -\lambda^{-1} \ln \int_0^\infty \mathcal{G}(t') e^{-\lambda t'} dt'. \quad (10)$$

This expression is exactly the same as that of the age of a radioactive tracer with decay λ and constant surface concentration, which can be seen by combining equations (1) and (8). Hence a conserved tracer with exponential growth yields the same age as a radioactive tracer that decays at the same rate.

[18] Expanding $e^{-\lambda t'}$ in equation (10) as a Taylor series about Γ we have [*Varni and Carrera*, 1998]

$$\tau = \Gamma - \lambda^{-1} \ln \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \lambda^n \mu_n, \quad (11)$$

where μ_n are the centered moments defined by equation (4). Neglecting terms of order higher than two we have

$$\tau \approx \Gamma - \lambda^{-1} \ln\left(1 + \lambda^2 \Delta^2\right). \quad (12)$$

If we further assume $\lambda\Delta \ll 1$, which corresponds to slow growth or decay of the tracer and/or a narrow TTD, this reduces to [*Hall and Plumb*, 1994]

$$\tau \approx \Gamma - \lambda \Delta^2. \quad (13)$$

Equations (12) and (13) show that the tracer age for a radioactive or exponentially increasing tracer is less than the mean age for nonzero Δ [see also *Deleersnijder et al.*, 2001]. In the limit of very slow growth or decay the tracer age equals the mean age, i.e., $\tau \to \Gamma$ as $\lambda \to 0$. The more rapid the growth or decay of the tracer the larger the sensitivity of τ to Δ, see Figure 3 below.

[19] The same approach as used above can be used to derive approximate formulae for τ_{He} in the idealized case where the surface tritium concentration decays exponentially. Making this assumption in equation (9) and again expanding exponential terms as Taylor series about Γ and retaining only up to second-order terms we have

$$\tau_{He} \approx \Gamma - \lambda^{-1} \ln\left(\frac{1 + (\lambda - \gamma)^2 \Delta^2}{1 + \gamma^2 \Delta^2}\right),$$

where γ is the decay rate of the surface concentration. Assuming $\gamma\Delta \ll 1$ and $|\lambda - \gamma|\,\Delta \ll 1$ this further reduces to

$$\tau_{He} \approx \Gamma - (\lambda - 2\gamma)\Delta^2.$$

In the limit $\gamma = 0$, i.e., constant surface concentrations, these equations reduce to equations (12) and (13), whereas if $\gamma = \lambda/2$ we have $\tau_{He} \approx \Gamma$. A more realistic limit is probably $\gamma \approx \lambda$, which corresponds to the surface concentration decaying at approximately the same rate as the tritium decay (this is the case for the tritium time series after 1970, see Figure 1 below). In this case $\tau_{He} \approx \Gamma + \lambda\Delta^2$ (for $\Delta \ll \lambda^{-1} \approx 20$ years); that is, the tritium-helium age is older than the mean age, with the difference increasing with the width of TTD. So for recent measurements in "young" waters, where the TTD samples only the decaying part of the tritium time series, the tritium-helium age is older than the mean age.

[20] As discussed in section 2, the ratio of two increasing tracers can also be used to define a tracer age. If the two tracers have exponential growth then the ratio of their surface concentrations will increase exponentially with growth rate equal to the difference in their growth rates; that is, if $a_0 = e^{\lambda_a t}$ and $b_0 = e^{\lambda_b t}$ then $R_0(t) = a_0(t)/b_0(t) = e^{(\lambda_a - \lambda_b)t}$. The above expressions (10)–(13) do not apply for the ratio age as the evolution equation satisfied by the ratio is not the same as the equation for individual tracers [*Doney et al.*, 1997], but it is still possible to relate the ratio age to the ages of the two individual exponentially increasing tracers. At an interior location where the concentration ages of the two tracers are τ_a and τ_b (i.e., $a(t) = e^{\lambda_a(t-\tau_a)}$ and $b(t) = e^{\lambda_b(t-\tau_b)}$) the time series of ratio of the tracers is given by

$$R(t) = \exp\{\lambda_b \delta_\tau - \delta\lambda(t - \tau_a)\}$$

where $\delta\tau = \tau_b - \tau_a$ and $\delta\lambda = \lambda_b - \lambda_a$. Using equation (7) to define the ratio age τ_R we then have

$$\tau_R - \tau_a = \frac{\lambda_b}{\delta\lambda} \delta t. \quad (14)$$

If the two tracers a and b have similar growth rates then $|\delta\lambda| \ll \lambda_b$ and the difference between the ratio age and either of the concentration ages will be much larger than the difference between the two concentration ages; that is, $|\tau_R - \tau_a| \gg \delta\tau$. Also, if $\lambda_a > \lambda_b$ then $\tau_R < \tau_a < \tau_b$.

[21] Finally, we consider idealized tracers that are conserved and whose surface concentration can be expressed as an order N polynomial in time. Writing $c_0(t)$ in the convolution (equation (1)) as a polynomial and using the definition for a concentration tracer age (equation (6)) it can be shown that

$$\sum_{n=1}^{N} \alpha_n (\tau^n - m_n) = 0, \quad (15)$$

where

$$\alpha_n(t) = (-1)^n \frac{1}{n!} \frac{d^n c_0}{dt^n}$$

and m_n are the moments of the TTD about the origin (equation (2)). Hence the tracer age of a tracer represented by a polynomial of order N depends only on the first N moments of the TTD. For linear growth ($N = 1$) equation (15) reduces to the above result that tracer age equals the mean age ($\tau = m_1 \equiv \Gamma$). For a tracer with quadratic temporal variation the tracer age depends only on the first two moments (mean age and width) of the TTD, and if the quadratic term is small equation (13) holds, with $\lambda = -2\alpha_2/\alpha_1$.

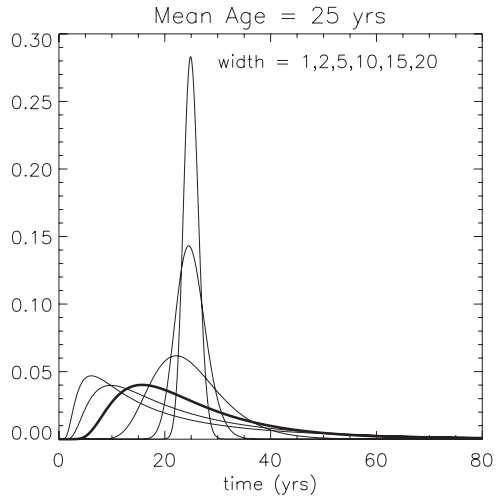


Figure 2. Inverse Gaussian transit time distributions with Γ = 25 years and Δ = 1, 2, 5, 10, 15, and 20 years.

[22] The above relationships for tracers with simple growth (or decay) illustrate that in general tracer ages will differ, and that the difference will depend on both the transient nature of the tracer and the characteristics of the TTD. However, it also suggests that knowledge of only the first two moments of the TTD may still be a very useful result for inferring the propagation of tracers with sufficiently weak growth or decay. Conversely, measurements of two tracers with linear and quadratic increases may tightly constrain the mean age and width of the TTD, see section 6 for further discussion.

4. Realistic Tracers

[23] We now examine the relationship among ages derived from different tracers for which measurements are available (in oceans, lakes, or groundwater). The approach used is to calculate hypothetical interior tracer time series $c(t)$ for each tracer for a specified transit time distribution $\mathcal{G}(t)$, and then to calculate the tracer age τ. Comparison of the tracer ages will show differences between the ages from different tracers, and also the sensitivity of the age to the characteristics of $\mathcal{G}(t)$. This sensitivity can also be used to examine what information about the TTD is contained in measurements of a particular tracer, and whether multiple tracers can be used to infer aspects of the TTD.

[24] In our analysis we focus primarily on TTD given by

$$\mathcal{G}(t) = \sqrt{\frac{\Gamma^3}{4\pi\Delta^2 t^3}} \exp\left(\frac{-\Gamma(t-\Gamma)^2}{4\Delta^2 t}\right). \qquad (16)$$

These TTD have a convenient form in terms of the mean age, Γ, and width, Δ, and by varying Γ and Δ a wide range of distributions can be formed. This includes near symmetric distributions ($\Delta \approx 0$) and asymmetric distributions with early peaks and long tails ($\Delta \approx \Gamma$), see Figure 2. Distributions of the form equation (16) are known as Inverse Gaussian (IG) distributions, and have been used in many different fields [e.g., *Chhikara and Folks*, 1989; *Seshadri*, 1999].

[25] The IG distribution is also the TTD for one-dimensional flow with constant velocity and diffusivity, with Γ^2/Δ^2 equal to the Peclet number [e.g., *Waugh and Hall*, 2002]. Tracer sensitivity to Γ and Δ could be equivalently described in terms of the Peclet number or the one-dimensional velocity, diffusivity and position [e.g., *Ekwurzel et al.*, 1994; *Sonnerup*, 2001]. However, we do not interpret the different Γ and Δ in our calculations in terms of one dimensional flows. We are using IG distributions simply because it has a convenient analytical form that enables a range of transit time distributions.

[26] In this section and the next we find, using the IG form, significant variation of tracer age with tracer and time. Were the real ocean TTD to have features not well reproduced by the IG form (e.g., bimodality) then even greater tracer and time variation would occur. A preliminary examination of the sensitivity of the tracer age calculations to the assumed functional form of the TTD is presented in section 7 below.

4.1. Surface Concentrations and Decay Rates

[27] We consider the following tracers: the halocarbons CFC-11, CFC-12, CFC-113, and CCl_4, their ratios, tritium ^{3}H (and daughter product ^{3}He), and also a family of tracers with constant surface concentrations but differing decay rates. The time-varying boundary conditions used in our calculations are shown in Figure 1. The halocarbon time series are from *Walker et al.* [2000], and in the calculations below we assume that halocarbon concentrations are at equilibrium (100% saturation) with the atmosphere in surface waters. Slightly different results will be obtained for different assumed saturation levels. The tritium time series is based on that by *Dreisigacker and Roether* [1978], and is appropriate for North Atlantic surface water. Again, different results will be obtained if a different time series is used.

[28] Figure 1a shows that the concentrations of all four halocarbons increased until the early 1990s. The rate of increase depends on the halocarbon and on time, see Table 1. The different growth rates mean that the ages from the different halocarbons will in general differ. It also means that ratios of the different halocarbons have changed. Figure 1b shows that the ratios CFC-11/CFC-12, CFC-11/CCl_4, and CFC-113/CFC-12 all increased monotonically between 1950 and (at least) 1980, and hence these can be used to define ages of water parcels formed during this period.

[29] As discussed in section 2, tritium ^{3}H and daughter product ^{3}He can be used to define a tracer age. We calculate the interior concentration of ^{3}H using equation (1) with decay rate $\lambda = 0.05576$ years^{-1} and the time series shown in Figure 1c as the surface concentration, while the interior concentration of ^{3}He equals the concentration of ^{3}H lost

Table 1. Approximate Exponential Growth Rate of Halocarbons Over 20 Year Periods[a]

Years	CFC-11	CFC-12	CFC-113	CCl_4
1950–1970	0.23	0.14	0.16	0.03
1955–1975	0.17	0.13	0.15	0.04
1960–1980	0.15	0.12	0.15	0.05
1965–1985	0.11	0.10	0.14	0.04
1970–1990	0.08	0.07	0.14	0.03
1975–1995	0.05	0.05	0.12	0.01

[a]Growth rates are in yr^{-1}.

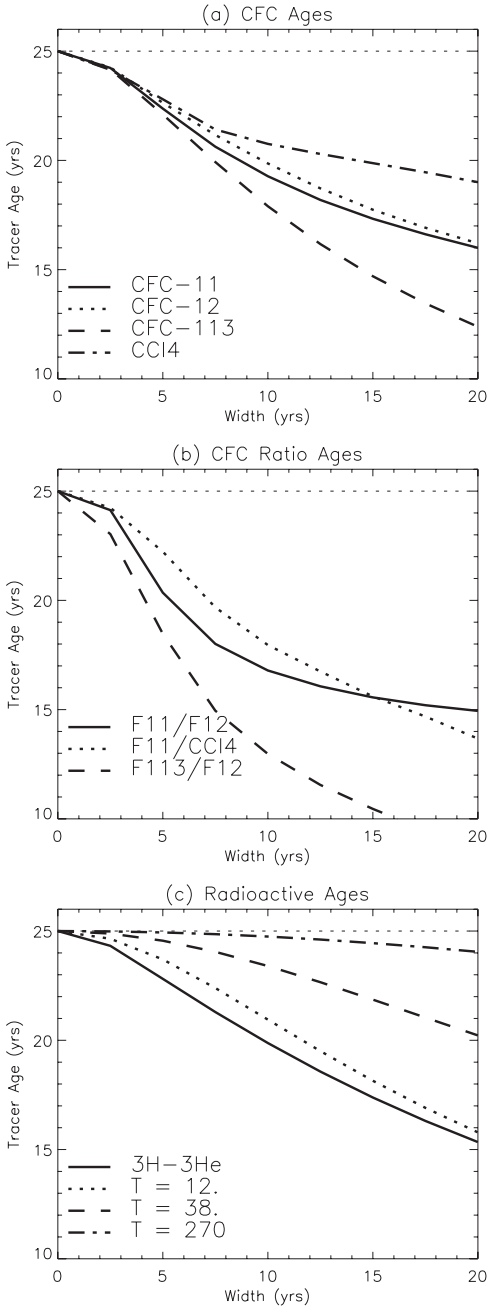


Figure 3. Variation of τ (in 1991) with width Δ for $\Gamma = 25$ years for different tracer ages: (a) halocarbon (CFC-11, CFC-12, CFC-113, and CCl_4) concentration ages,(b) halocarbon ratio (CFC-11/CFC-12, CFC-113/CFC-12, and CFC-11/CCl_4) ages, and (c) tritium-helium and radioactive tracer ages. Note that y axes cover only 10–26 years.

through radioactive decay. These concentrations are then used in equation (9) to calculate the tritium-helium age.

[30] Calculations have also been performed for several tracers with constant surface concentrations but differing decay rates. Included in these calculations are tracers with decay rates similar to tritium, argon 39 (half-life $T = 269$ years), and carbon 14 ($T = 5370$ years). We also consider a radioactive tracer with decay rate similar to the growth rate of CO_2 in the atmosphere ($T = 38$ years). As discussed in section 3, the age of a radioactive tracer equals the surface interior concentration time lag of conserved tracer with exponential growth at same rate as radioactive decay. Therefore the age of radioactive tracer with $T = 38$ years will be very similar to the surface interior time lag of anthropogenic carbon in the ocean (assuming the anthropogenic carbon biogeochemistry is linear). This means that the comparison of the age from a radioactive tracer with $T = 38$ years and the other tracer ages will provide insight into how well different tracer ages approximate the time lag of anthropogenic carbon in the ocean.

4.2. Illustrative Examples

[31] Before examining the age-age relationships for a wide range of TTDs, we first present example calculations that illustrate the differences between the tracer ages and the sensitivity of tracer ages to the TTD. Figure 3 shows the variation of different tracer ages (in 1991) with width Δ, for mean age $\Gamma = 25$ years. For $\Delta = 0$ all tracer ages equal the mean age, but for $\Delta > 0$ the tracer ages are younger than the mean age and different tracers yield different ages. The larger Δ, corresponding to more mixing, the larger the difference between tracer ages.

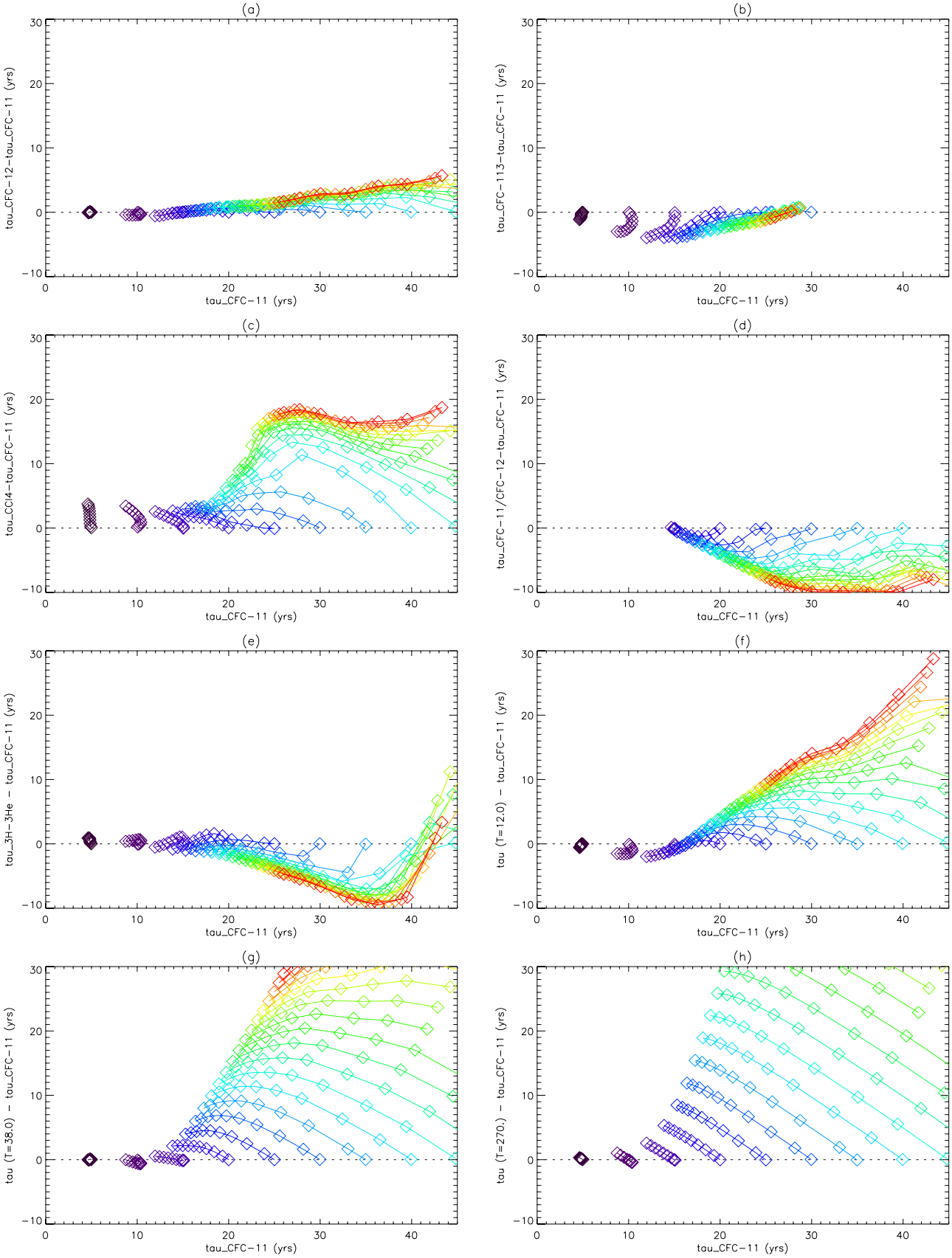
[32] The differences between the halocarbon ages shown in Figure 3a can be understood in terms of the differing growth rates and the analysis in section 3. For the 20-year period before 1991 CFC-113 had the fastest growth rate, CCl_4 the slowest, and CFC-11 and CFC-12 had similar growth rates (Table 1). Thus $\tau_{CFC113} < \tau_{CFC11} < \tau_{CFC12} < \tau_{CCl4}$ (where τ_{CFC113} is the tracer age from CFC-113), with small differences between τ_{CFC11} and τ_{CFC12}. A similar dependence is seen in Figure 3c where there is larger sensitivity to Δ for larger radioactive decay, again consistent with the approximate age equation (13).

4.3. Age-Age Relationships

[33] We now examine the interrelationship of tracer ages over a range of TTDs. We show calculations of tracer ages in 1991, but as discussed in the next section there can be significant temporal variations in tracer ages. Calculations similar to those presented below have been performed for other years, and there are quantitative differences but the qualitative features are the same.

[34] Figure 4 shows the relationship, in 1991, of the ages calculated from several different tracers with the age from CFC-11. These plots clearly show that the age-age relationships depend on both the tracers and the TTD. As discussed in the previous subsection, the differences in the growth rates of the halocarbons over the 1970s and 1980s means that $\tau_{CFC113} \leq \tau_{CFC11} \leq \tau_{CFC12} \leq \tau_{CCl4}$. Figure 4 shows that over a wide range of TTD the differences between

Figure 4. (opposite) Relationship between $\delta\tau_X = \tau_X - \tau_{CFC11}$ and τ_{CFC11} (in 1991) for several different tracer ages τ_X: (a) CFC-12, (b) CFC-113, (c) CCl_4, (d) CFC-11/CFC-12, (e) ^3H-^3He, and (f)–(h) radioactive tracers with $T = 12$, 38, and 270 years. The different curves correspond to different mean ages ($\Gamma = 5$, 10,.., 100 years), and symbols correspond to the different widths (Δ varies from 0 to Γ in 10 equal intervals). The symbols on the line $\delta\tau_X = 0$ correspond to $\Delta = 0$, whereas those at the end of the curves correspond to $\Delta = \Gamma$.


(a)
tau_CFC-12-tau_CFC-11 (yrs)
tau_CFC-11 (yrs)
(b)
tau_CFC-113-tau_CFC-11 (yrs)
tau_CFC-11 (yrs)
(c)
tau_CCl4-tau_CFC-11 (yrs)
tau_CFC-11 (yrs)
(d)
tau_CFC-11/CFC-12-tau_CFC-11 (yrs)
tau_CFC-11 (yrs)
(e)
tau_3H-3He - tau_CFC-11 (yrs)
tau_CFC-11 (yrs)
(f)
tau (T=12.0) - tau_CFC-11 (yrs)
tau_CFC-11 (yrs)
(g)
tau (T=38.0) - tau_CFC-11 (yrs)
tau_CFC-11 (yrs)
(h)
tau (T=270.) - tau_CFC-11 (yrs)
tau_CFC-11 (yrs)

τ_{CFC12} and τ_{CFC11} are small, with differences larger than 2 years only for $\tau_{CFC11} > 25$ years [see also *Sonnerup*, 2001]. There are also small differences between τ_{CFC113} and τ_{CFC11}, except now the largest differences occur for $\tau_{CFC11} \approx 15$ years, where τ_{CFC113} can be as young as 10 years. In contrast to the above CFCs, $\tau_{CCl4} > \tau_{CFC11}$ for all finite width TTD, with differences as large as 15 years possible for $\tau_{CFC11} > 25$ years.

[35] The CFC11/CFC12 ratio age $\tau_{11/12}$ is less than τ_{CFC11} for all TTD with nonzero width, and the magnitude of the difference is, in general, much larger than between τ_{CFC12} and τ_{CFC11}, see Figure 4d [see also *Sonnerup*, 2001]. This is consistent with the discussion in section 3: Substituting the approximate growth rates for CFC-11 and CFC-12 over the period 1970 to 1990 (0.08 and 0.07 respectively; see Table 1) into equation (14) we have $\delta\tau_{11/12} \approx -7\ \delta\tau_{CFC12}$.

[36] There are only small differences between the tritium-helium age τ_{He} and τ_{CFC11} for $\tau_{CFC11} < 15$ years, but τ_{He} can be less than τ_{CFC11} by up to 10 years for older τ_{CFC11}, see Figure 4e [see also *Doney et al.*, 1997]. Although there can be large differences between τ_{He} and τ_{CFC11}, it is notable that the τ_{He} and τ_{CFC11} relationship is fairly compact (especially if TTD with $\Delta \approx 0$ are excluded). This indicates that the $\tau_{He} - \tau_{CFC11}$ relationship is relatively insensitive to changes in the flow, as noted by *Doney et al.* [1997]. This insensitivity limits the amount of information on the TTD that can be obtained from simultaneous measurements of CFC-11 and tritium-helium (see section 6).

[37] The ages from all three radioactive tracers differ significantly from τ_{CFC11} for $\tau_{CFC11} > 15$ years, with increasing difference for decreasing decay rates (consistent with analysis in section 3), see Figures 4f–4h. As discussed above the tracer age from a radioactive tracer with T = 38 years approximates the time lag in concentration of anthropogenic carbon (τ_{CO2}), and so Figure 4g shows the relationship of τ_{CFC11} with τ_{CO2}. Understanding this relationship is important as the CFC-11 age has been used in several studies to calculate carbon uptake [e.g., *Gruber*, 1998; *Sabine et al.*, 1999]. Although there is good agreement for $\tau_{CFC11} < 15$ years, τ_{CO2} can be much greater than τ_{CFC11} for larger ages, for example, for flows with $\tau_{CFC11} \approx 25$ years τ_{CO2} can be as large as 45 years. Because of this there can be a large bias in lower thermocline estimates of anthropogenic carbon that use CFC ages to infer the time lag in anthropogenic carbon (see *Hall et al.* [2002] for further discussion).

[38] In the above analysis we have focused on decadal-scale transport scales. We now briefly discuss longer timescales (>100 years), which are more appropriate for the deep ocean. In the deep ocean there is very little, if any, of the anthropogenic tracers, and the most useful tracers are radioactive tracer with half-lives greater than 100 years. Figure 5 shows the relationship between two such tracers τ_{39Ar} (half-life of 269 years) and τ_{14C} (half-life of 5370 years), for a range of Γ and Δ. See *Broecker and Peng* [2000] for a similar $\tau_{39Ar} - \tau_{14C}$ plot for observations. For the range of values shown $\tau_{14C} \approx \Gamma$ as the decay of ^{14}C is approximately linear over these timescales. However, because of its more rapid decay, there is a large decrease in τ_{39Ar} with increasing Δ. So even with only moderate mixing the two radioactive ages differ. As discussed in section 6 this suggests that joint measurements of these tracers may constrain the TTD in the deep ocean.

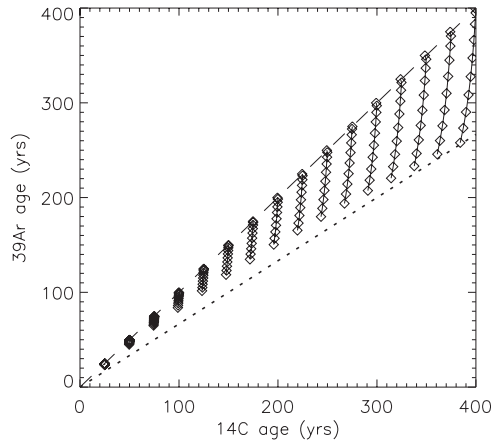


Figure 5. Relationship of between τ_{14C} and τ_{39Ar} for TTD with Γ from 25 to 400 years. As in Figure 4 each curve corresponds to constant Γ and the symbols to Δ varying from 0 to Γ in equal increments. The dashed line corresponds to $\tau_{14C} = \tau_{39Ar}$, and the dotted line corresponds to $\tau_{14C} = 1.5\ \tau_{39Ar}$.

5. Temporal Variations of Tracer Ages

[39] In the above calculations all ages were calculated in 1991. However, for many tracers the age varies with time even if the transport is steady. This is illustrated in Figure 6, which shows the temporal variation of tracer age for an TTD with Γ = 25 years and Δ = 10 years for the same tracers as in Figure 3. This shows that the ages calculated from different tracers vary with analysis (measurement) date, except for radioactive tracers with constant surface concentrations. The temporal variation in the halocarbon tracer ages is consistent with the temporal changes in growth rates of the tracers. The CFC-113 growth rate was relatively constant after 1960 and τ_{CFC113} is fairly constant with time, whereas the growth rates of CFC-11 and CFC-12 decreased with time and τ_{CFC11} and τ_{CFC12} increase with time. There are also significant temporal variations in the tritium-helium age, and this age is generally very different from the age of a tracer with same decay rate (T = 12.43 years) but constant surface concentration.

[40] We now focus on the CFC-11 and tritium-helium ages, and examine the sensitivity of the time dependence to the characteristics of the TTD. Figure 7 shows the temporal evolution of τ_{CFC11} and τ_{He} for several different TTD. Each plot shows three sets of curves, where each set of curves correspond to different TTD for which the tracer age equals a fixed value τ_0(= 10, 15, or 20 years) in 1985. For advective flow (Δ = 0, dotted lines) the tracer ages are independent of analysis date. However, for finite width TTD there are significant temporal variations in both tracer ages, especially over the last two decades, when the ages increase with analysis date.

[41] It is notable in Figure 7 that the temporal increase in age is relatively insensitive to width of the TTD. This suggest that temporal changes that occur under steady

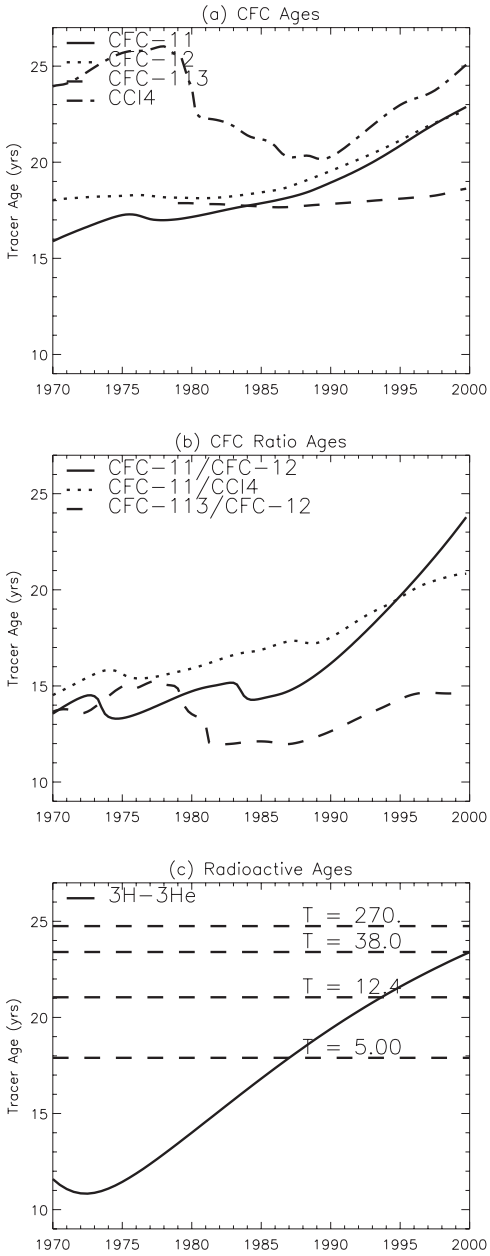


Figure 6. Temporal variation of τ for TTD with $\Gamma = 25$ years and $\Delta = 10$ years for the same tracer ages as shown in Figure 3. τ_{CFC113} is not shown before 1980 because the interior concentration of CFC-113 is negligible for these dates.

transport can be examined without detailed knowledge of the TTD. However, it also means that repeated measurements of these transient tracers may not yield much information on the width of the TTD.

[42] The above calculations of the temporal variations in tracer ages have important implications for inferring changes in ocean ventilation from temporal changes in tracer ages, as has been done in several recent studies [e.g., *Doney et al.*, 1998; *Roether et al.*, 1998; *Min et al.*, 2000; *Watanabe et al.*, 2001]. Before attributing changes in ages to temporal changes in the transport it is necessary to know what temporal changes in tracer ages can occur even when there is steady transport. We now briefly compare the above calculations with the observed changes in tracer ages.

[43] *Robbins and Jenkins* [1998] analyzed τ_{He} from measurements in the eastern North Atlantic subtropical gyre between 1979 and 1993, and showed a significant increase in the age, especially for the locations with older ages. For example, τ_{He} on the $\sigma_\theta = 27.1$ kg m^{-3} isopycnal surface at 30W, 26N increased from around 10 to 16 years over this period. This increase is very similar to that shown in Figure 7, for moderate to large mixing. Hence, as was clearly stated by *Robbins and Jenkins* [1998], the observed change in tracer age are not inconsistent with steady transport, and so the data does not imply a change in transport. Changes in τ_{CFC12} in the eastern North Atlantic have also been observed. For example, *Doney et al.* [1998] showed increases from around 15–20 years to 18–24 years in the subtropical gyre between 1988 and 1993. These changes are again similar to those shown in Figure 7, and do not necessarily imply changes in the circulation.

[44] *Watanabe et al.* [2001] analyzed changes in τ_{CFC11} in the North Pacific between 1985–1987 and 1999–2000, and showed an increase of around 10 years, for water masses with ages around 15 years in 1985. They attributed this change to changes in water formation. However, the calculations in Figure 7 show that changes as large as 7

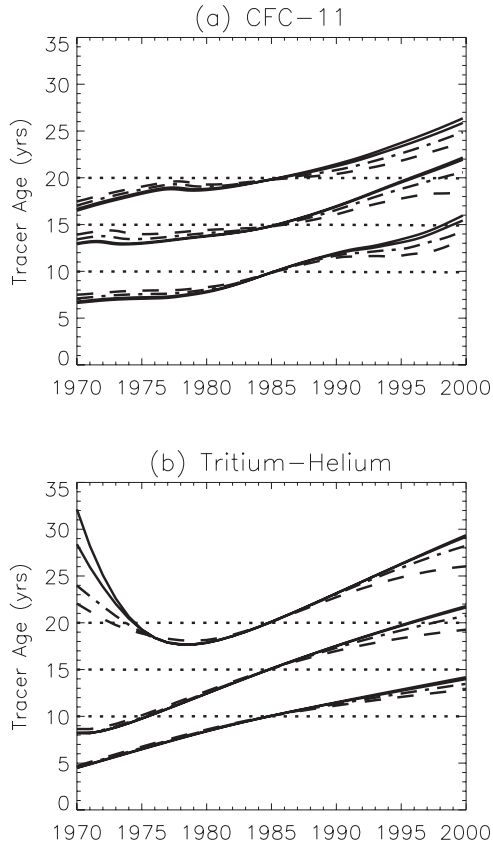


Figure 7. Temporal variation of (a) τ_{CFC11} and (b) τ_{He}. Each set of curves has the same tracer age in 1985 ($\tau_0 = 10$, 15, and 20 years) but varying TTD. Within each set, $\Gamma = (1, 1.25, 1.5, 2, 2.5)\tau_0$, and Δ is determined so that the tracer age equals τ_0 in 1985 ($\Delta = 0$ for $\Gamma = \tau_0$).

years can be caused by steady transport. So although steady transport (with TTD similar to an IG distribution) cannot explain all the increase it can explain a large fraction of it, and attributing all the change to transport may be erroneous.

[45] Conversely, inferring constant transport from constant tracer ages may also be erroneous. For example, *Min et al.* [2000] report remarkably little change in τ_{CFC11} in the Eastern North Pacific between 1982 and 1994. However, from the calculations above we would expect an increase of around 6 years if steady transport, with moderate to large mixing. The lack of change in observed ages is consistent with nearly advective transport as well as transport with mixing, where the transport rate increases in time to cancel the tendency for tracer age to increase.

[46] In summary, the above calculations show that significant temporal changes in CFC and tritium-helium tracer ages can occur even when there is steady transport, and these changes are of similar magnitude to observed changes in the North Pacific and North Atlantic. Hence great care needs to be taken when using changes in tracer ages to infer temporal changes in the circulation.

6. Constraining the Transit Time Distribution

[47] The calculations shown in section 4 show that there can be significant differences between ages derived from different tracers. While this complicates the interpretation of single age measurements, it does raise the possibility of using tracer ages in combination to constrain the TTD. Here, we examine whether ages from two tracers can be used to determine the first two moments of the TTD (i.e., the mean age and width). In a concurrent study, the concentrations of tracer instead of tracer ages are used to constrain the TTD and estimate the uptake of anthropogenic carbon [*Hall et al.*, 2002].

[48] Assuming the TTD is the same form as the IG distribution, a given tracer age constrains Γ and Δ to a range of values (with $\Gamma = \tau$ and $\Delta = 0$ being one limit). This can be seen in Figure 4, where there is a range of Γ (curves) which yield a fixed value of τ_{CFC11}. A clearer way to see this are contour plots of τ for varying Γ and Δ. Figure 8 shows such plots for the tracers shown in Figure 4 (in each plot the dashed contours correspond to contours of τ_{CFC11}). In each case, a given tracer age corresponds to a range of mean age and width, i.e., measurement of a tracer age constrain Γ and Δ along that age contour in the plots in Figure 8. The smallest Γ for given tracer age corresponds to Γ equal to the tracer age (with $\Delta = 0$). TTD with larger values of Γ produce the same tracer age if the corresponding Δ are also larger. Progressively larger (Γ, Δ) values correspond to increasingly asymmetric IG distributions. These TTD represent water masses that have young components with high tracer concentration mixed with old components with low concentration such that the "observed" tracer age is matched.

[49] In Figure 8 the TTD is assumed to have an IG distribution. The tracer ages, and hence contours, may be sensitive to this assumption for the largest Γ and Δ shown. For these old TTDs the transient tracers have sampled only a small fraction of the TTD and their ages provide no information on the characteristics of the older part of the TTD. For younger Γ and Δ the tracers have fully sampled the TTD and their ages will be less sensitive to the shape of the TTD. See next section for further discussion.

[50] We consider now whether the addition of a second tracer age will reduce the possible Γ and Δ for given measurement of τ_{CFC11}. We first consider addition of measurements of τ_{CFC12}. For most of the domain in Figure 8a the τ_{CFC11} and τ_{CFC12} contours are very similar and measurements of the two tracer ages will not constrain the TTD much more (if at all) than a single age measurement. The exception is for $\Gamma > 30$ years and $\Delta < 10$ years where the τ_{CFC11} and τ_{CFC12} contours have different slopes and the two measurements, of sufficient accuracy, may provide addition constraints. For example, if the tracer ages differ by more than around two years lower bounds on Γ and Δ may be determined, with Γ greater than the tracer ages and Δ greater then zero. This eliminates the possibility of purely advective flow. Conversely, if the tracer ages are equal (and older then 30 years) this implies advective flow.

[51] The situation for τ_{He} is similar to that for τ_{CFC12}. For young ages ($\tau_{CFC11} < 20$ years), the addition of τ_{He} measurements will not likely constrain Γ and Δ more than just measurements of τ_{CFC11}, whereas for older ages the range of possible Γ and Δ may be constrained. The constraints from joint τ_{He} and τ_{CFC11} measurements are tighter than those by τ_{CFC11} and τ_{CFC12} measurements, and in some situations it may be possible to put both upper and lower limits on Γ and Δ. For example, measurements of $\tau_{He} = 25$ years and $\tau_{CFC11} = 30$ years, with uncertainties of 1 year, constrain (Γ, Δ) to be between around (35, 5) and (75, 40) years.

[52] Although not measured as often as the above tracers, measurements of CFC-113 or CCl_4 together with CFC-11 may be very useful for constraining the characteristics of the TTD. The age contours in Figure 8 for these tracers are not parallel for young of tracer ages ($\tau_{CFC11} < 15$ years), and measurements of either pair in young water masses may reduce the range of possible (Γ, Δ). For example, measurements of $\tau_{CFC11} = 10$ years and $\tau_{CCl4} = 12$ years imply $\Gamma \approx 11$ years and $\Delta \approx 6$ years, while a measurement of $\tau_{CFC113} = 8$ years with the same τ_{CFC11} measurement yields a similar constraint on Γ and Δ. How tightly constrained the Γ and Δ are around this values depends on the measurement uncertainties. For age uncertainties of 1 year, Γ is constrained within 3 years of the above value.

[53] The tightness of the constraint also varies with the values of τ_{CCl4} or τ_{CFC113}, for the same value of τ_{CFC11}. In general, the closer τ_{CFC113} or τ_{CCl4} are to τ_{CFC11} the tighter the constraint, with Γ constrained close to the tracer ages and Δ close to zero. For larger differences from τ_{CFC11} (i.e., larger τ_{CCl4} or smaller τ_{CFC113}) there is generally a larger range of possible values of Γ and Δ, with possibly only lower limits. However, even though a wide range of TTD is possible, in this case it is notable that narrow TTD, and hence advective flows, are eliminated.

[54] Another transient tracer that we have not considered so far is sulfur hexafluoride, SF_6. The atmospheric concentration of SF_6 has increased rapidly and monotonically in the last two decades [e.g., *Maiss et al.*, 1996] and it can be used to define a tracer age [e.g., *Vollmer and Weiss*, 2002]. Calculations (not shown) indicate that measurements of

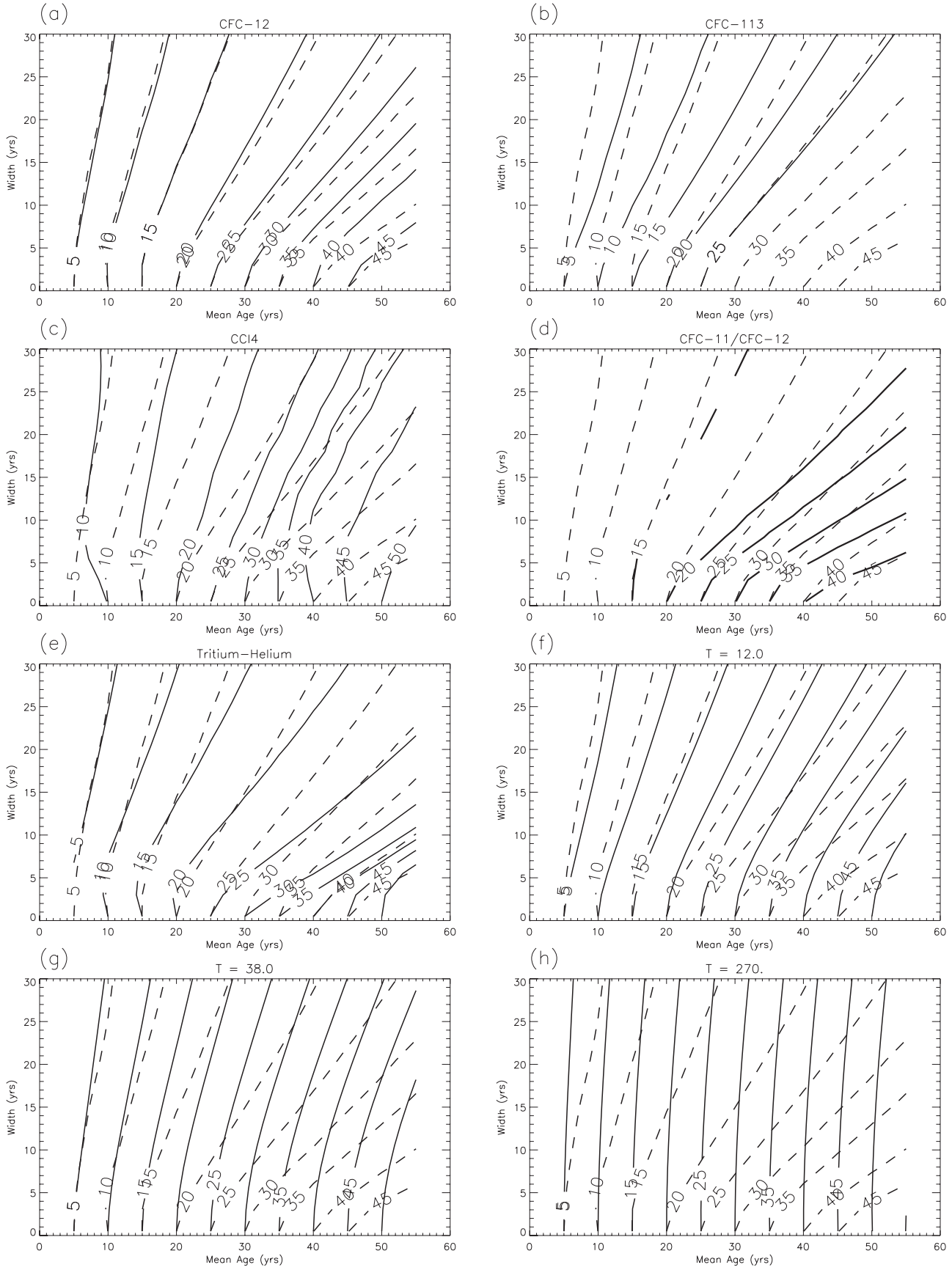


Figure 8. Variation of tracer ages (in 1991) with Γ and Δ for (a) CFC-12, (b) CFC-113, (c) CCl_4, (d) CFC-11/CFC-12, (e) ^3H-^3He, and (f)–(h) radioactive tracers with $T = 12$, 38, and 270 years. In each plot the dashed contours correspond to τ_{CFC11}. Tracer age contours for 5, 10, 15,..., 50 years are shown.

τ_{SF6} in young water masses will provide similar constraints on the TTD to those by τ_{CFC113}, and so SF_6 could also be a very useful tracer.

[55] For the older waters ($\tau_{CFC11} > 25$ years) a CFC-113 (or SF_6) age cannot be defined. However, a CCl_4 age can be calculated, and again τ_{CCl4} together with τ_{CFC11} may tightly constrain Γ and Δ. For example, measurements of τ_{CFC11} = 30 years and τ_{CCl4} = 45 years imply $\Gamma \approx 54$ years and $\Delta \approx$ 24 years (see Figure 8c).

[56] In the calculations shown we assumed that all halocarbons are conserved, but several recent studies have shown that this is not always the case for CFC-113 [*Roether et al.*, 2001] and CCl_4 [e.g., *Wallace et al.*, 1994; *Meredith et al.*, 1996; *Huhn et al.*, 2001]. The existence of an undetermined degradation of CFC-113 and CCl_4 will limit their use as a quantitative tracer. However, if the degradation rates can be accurately determined it would be possible to include a decay term in the TTD analysis (see equation (1)), and in the calculation of tracer ages.

[57] We finally consider measurements of the radioactive tracers ^{39}Ar and ^{14}C. Although there are large uncertainties in the age calculated from these measurements and there are currently limited measurements of ^{39}Ar, these measurements may be useful in constraining the TTD in older waters. For the timescales relevant for water with $\tau_{CFC11} \approx 30$ years τ_{39Ar} and τ_{14C} approximate Γ, and measurements of these ages alone will constrain Γ to similar values as the age calculation. If measurements of one of these radioactive tracers are combined with one of the above anthropogenic tracers it may then be possible to also constrain Δ. For example, a measurement of $\tau_{39Ar} = 45 \pm 5$ years together with $\tau_{CFC11} = 30 \pm 1$ years constrain (Γ, Δ) between around (40, 10) and (50, 20) years.

[58] For older waters than considered above, where the halocarbons have not yet penetrated, simultaneous measurements of τ_{39Ar} and τ_{14C} may constrain Γ and Δ. As shown in Figure 5, for τ_{39Ar} and τ_{14C} in the range of 100 to 400 years $\tau_{14C} \approx \Gamma$ whereas τ_{39Ar} is much less than Γ for moderate to large Δ. So measurements of τ_{14C} will constrain Γ, while the addition of τ_{39Ar} measurements will enable Δ to be constrained. For example, if $\tau_{14C} = 200 \pm 10$ years and $\tau_{39Ar} = 150 \pm 10$ years then $190 < \Gamma < 220$ years and $130 < \Delta < 280$ years.

[59] Although a detailed analysis of actual measurements of tracer ages is beyond the scope of this study, we briefly discuss the constraints on the TTD imposed by some published age measurements.

[60] *Sonnerup* [2001] compared τ_{CFC11}, τ_{CFC12}, and $\tau_{11/12}$ measurements in the North Pacific Ocean [see also *Warner et al.*, 1996]. Consistent with the above discussion there are very similar CFC ages for water with $\tau_{CFC11} < 20$ years, but for older water τ_{CFC12} is older than τ_{CFC11} by up to 4 years and $\tau_{11/12}$ is less than τ_{CFC11} by up to 10 years. These variations are consistent with Figure 4, with the observed age-age relations being near the maximum differences in Figure 4. *Sonnerup* [2001] reported values of $\tau_{CFC11} = 29 \pm 0.4$ years, $\tau_{CFC12} = 31.4 \pm 0.4$ years, and $\tau_{11/12} = 20 \pm 3$ years for measurements at 650m and (152W, 34N). Applying the above analysis to these measurements constrains Γ and Δ to be greater than 45 and 15 years, respectively, which corresponds to a large range of transit times from the surface to the above location.

[61] We have performed preliminary analysis of CFC-11 and CFC-12 measurements (from WOCE) in the Atlantic and Indian Oceans, and the $\tau_{CFC11} - \tau_{CFC12}$ relationships are very similar to those shown by *Sonnerup* [2001]; that is, for $\tau_{CFC11} \approx 30$ years τ_{CFC12} is older by 2 to 3 years. So in all these oceans water masses with $\tau_{CFC11} > 20$ years have broad TTD, with mean age much older than the CFC tracer ages. This may also be true for younger τ_{CFC11} ages but the joint analysis of τ_{CFC11} and τ_{CFC12} alone cannot distinguish between broad or narrow TTD.

[62] Simultaneous measurements of τ_{He} and τ_{CFC12} in the North Atlantic Ocean have been reported by *Doney et al.* [1997]. They report similar values of τ_{CFC12} and τ_{He} for ages less than 15 years, but for older ages $\tau_{He} < \tau_{CFC12}$ with the difference as large as 10 years for $\tau_{CFC12} \approx 30$ years. The measurements in the younger water do not constrain the TTD much more than a single tracer age, however the measurements in the older water do provide some constraint. For example, $\tau_{CFC12} \approx 30$ years and $\tau_{He} \approx 20$ years in 1988 imply $\Gamma > 60$ years and $\Delta >$ 30 years, i.e., very broad TTD with mean age at least twice τ_{CFC12}.

[63] As mentioned above, another useful transient tracer is sulfur hexafluoride (SF_6). *Vollmer and Weiss* [2002] recently reported ages from measurements of CFC-11, CFC-12 and SF_6 in the North Pacific Ocean (44N, 125W). At shallow depths (100–300 m) the CFC ages were very similar (increasing from 18 to 24 years) but the SF_6 ages were younger by 4–5 years. These measurements provide lower limits for Γ and Δ, with $\Gamma > 20$ years and $\Delta >$ 15 years for the youngest water mass. For the deepest waters sampled an SF_6 age cannot be calculated but CFC ages can be calculated and, consistent with the above CFC measurements in the North Pacific, the τ_{CFC11} is younger than τ_{CFC12} by around 3 years, indicating that $\Gamma > 55$ years and $\Delta > 15$ years.

[64] Transient tracers have been also been measured in lakes. *Vollmer et al.* [2002] recently reported measurements of the three CFCs, SF_6, tritium and helium in Lake Issyk-Kul. The different tracers yield different ages, with the values at the bottom of the lake varying between 9 and 13 years. Preliminary analysis indicates that these measurements tightly constrain Γ and Δ, with (Γ, Δ) $\approx$ (10, 5) years for the deepest measurements. These values again correspond to broad TTD, with transit times ranging from values shorter than the tracer ages to values larger than the tracer ages. A more detailed analysis of these data is currently underway.

[65] *Broecker and Peng* [2000] compared observed ages from ^{39}Ar and ^{14}C in the deep oceans. They show that on average τ_{14C} is 1.5 times τ_{39Ar}. Figure 5 indicates that TTD with $\Delta > \Gamma$ are required to produce $\tau_{14C} = 1.5\ \tau_{39Ar}$, which suggests that the TTD in the deep oceans are very broad with widths comparable to their mean ages.

7. Sensitivity to Shape of the Transit Time Distribution

[66] In the above calculations all TTD are of the form equation (16). This is not a major issue for the discussions

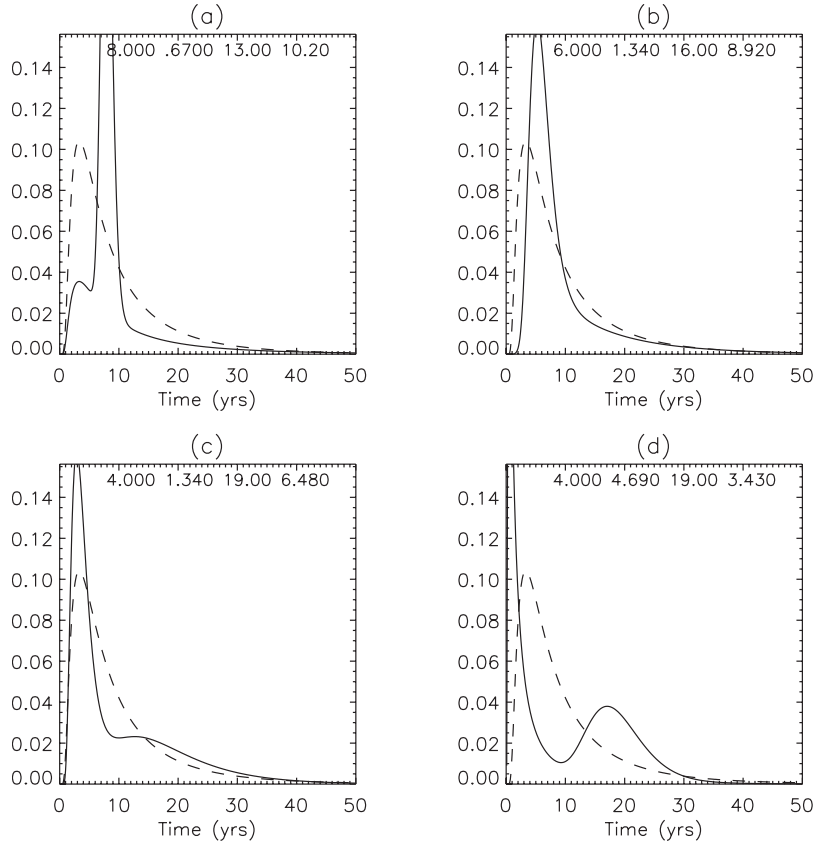


Figure 9. TTD within the family of two-IG distributions, as defined in the text, with $\Gamma = 10$ years and $\Delta = 6.7$ years (the dashed curves are the single IG TTD with the same age and width). In each panel the values of Γ_1, Δ_1, Γ_2, and Δ_2 are listed ($\alpha = 0.6$ in all panels).

in sections 4 and 5, as we were primarily interested in the range of variations of the ages (between tracers or with time) and the family of IG distribution covers a wide range of TTD shapes. However, using IG distributions could have a large impact on the inferences of Γ and Δ from multiple tracer ages, especially when the tracers have only sampled a small fraction of the TTD.

[67] While the TTD in multidimensional model simulations generally have the same characteristics as the IG distribution (broad, asymmetric distributions with a single early peak and a long tail), bimodal TTD can be seen in some flow simulations [e.g., *Haine and Hall*, 2002; A. Putzka, unpublished manuscript, 1999]. The possibility of such TTD should therefore be considered when trying to infer Γ and Δ from multiple tracer ages discussed in the previous section. A full examination of the sensitivity of tracer ages to shape of the TTD is beyond the scope of this study. However we present below a preliminary examination of this sensitivity.

[68] We examine the sensitivity by considering the variation of the tracer ages within families of TTD with the same Γ and Δ but differing shapes. This provides insight into the sensitivity of inferred (Γ, Δ) from the multiple tracer analysis to the assumed shape of the TTD. If for fixed (Γ, Δ) there is little sensitivity of the tracer ages to the shape of the TTD then we can have confidence in the inferred values of Γ and Δ, but not necessarily the shape, from the multiple age analysis.

[69] In this analysis we consider TTD that are a linear combination of two IG distributions, i.e.,

$$\mathcal{G} = \alpha\mathcal{G}_1(\Gamma_1, \Delta_1) + (1 - \alpha)\mathcal{G}_2(\Gamma_2, \Delta_2) \tag{17}$$

where $\mathcal{G}_i$ ($i = 1, 2$) are IG functions given by equation (16) with mean ages and widths Γ_i and Δ_i and $0 < \alpha \leq 1$. A linear combination of two IG distributions has been used previously in the oceanic (Putzka, unpublished manuscript, 1999) and atmospheric [*Andrews et al.*, 2001] context. Simple analytical expressions can be derived which give the mean age Γ and width Δ of this TTD in terms of α, Γ_i and Δ_i.

[70] For given mean age Γ and width Δ we have calculated tracer ages for a large number of the above "two-IG" distributions. Specifically, we varied α from 0.1 to 0.9, Γ_1 from 0.1 to 0.9 times Γ, and Δ_1 from 0.1 to 0.9 times Δ, and for each combination of these 3 parameters we determined Γ_2 and Δ_2 so that the mean and width of TTD equaled Γ and Δ. (Note that for around half the 1000 combinations of (α, Γ_1, Δ_1) it is not possible to form TTD with given Γ and Δ.)

[71] Figure 9 shows four different TTD within the family of two-IG TTD with $\Gamma = 10$ years and $\Delta = 6.7$ years. This plot shows that there can be considerable variations in the shape of the TTD even though the mean and width are fixed. The TTD can be unimodal or bimodal, and in the bimodal case the smaller peak can occur before or after the

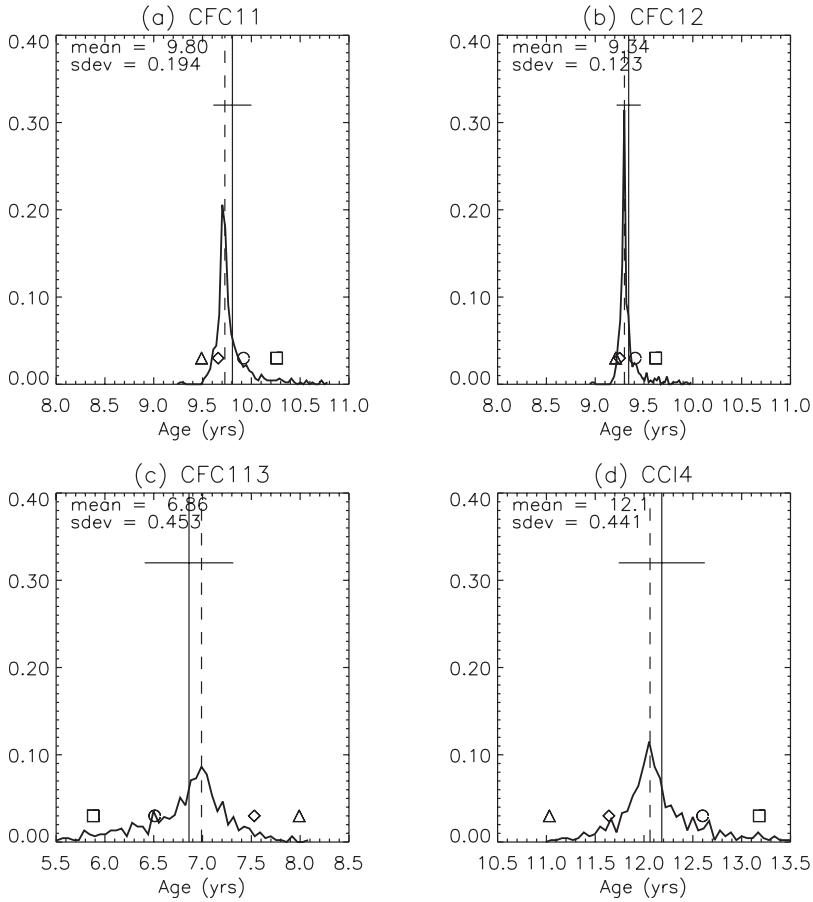


Figure 10. Distributions of tracer ages for all TTD within the family of two-IG distributions with Γ = 10 years and Δ = 6.7 years for (a) CFC-11, (b) CFC-12, (c) CFC-113, and (d) CCl_4. The vertical line is the mean value over the distribution, the horizontal line is ±1 standard deviation about this mean, and the vertical dashed line is the value for the single IG TTD. The triangles, diamonds, circles, and squares mark the tracer ages for the TTD shown in Figures 8a, 8b, 8c, and 8d, respectively. Note the different x axes in the different panels.

larger peak. Furthermore the location of the largest peak, and hence the most common transit time, can vary from 1 to 9 years.

[72] The variations in the TTD shape result in variations in the tracer ages. The variation of τ_{CFC11}, τ_{CFC12}, τ_{CFC113}, and τ_{CCl4} over the whole distribution of two-IG TTD with Γ = 10 years and Δ = 6.7 years is shown in Figure 10. The ages for the four TTD shown in Figure 9 are also shown (symbols). Figure 10 shows that the halocarbon tracer ages are relatively insensitive to the exact details of the TTD. Although there can be large differences in the shape of the TTD (e.g., Figure 9) the variation in the tracer ages is small (and probably less than the uncertainty in age measurements). This means that the restriction to IG TTD does not have a large impact on the inferences of Γ and Δ from multiple tracer ages, for "young" TTD.

[73] This insensitivity can be understood by returning to the analysis in section 3 where expressions for tracer ages involving only Γ and Δ were derived for tracers with quadratic growth. This means that the tracer age of a tracer with near quadratic growth over the history of the TTD is insensitive to third and higher moments of the TTD. For the time period sampled by TTD with Γ = 10 years and Δ = 6.7 years (1960–1990) quadratic growth is a reasonable approximation for the halocarbons, and so the halocarbon ages are relatively insensitive to higher-order moments of the TTD (and the existence of one or two modes in the TTD).

[74] We have performed the above calculations for other families of two-IG TTD, with a range of Γ and Δ. Figure 11 shows the variation of the mean and standard deviation of the tracer age distributions with Γ, for Γ varying from 10 to 60 years and $\Delta = 2\,\Gamma/3$. This shows that both the tracer age and the sensitivity of the age to the shape (i.e., standard deviation of the distributions) increase with the age of the TTD. The sensitivity is increased because for older mean ages there is a large fraction of the TTD that has not been sampled by halocarbons (i.e., for 1991 measurements there is no CFC-113 in the fraction of the TTD older than 30 years), and so the halocarbons provide no information on the old part of the TTD.

[75] In cases where a large fraction of the TTD has not been sampled by the transient tracers the calculations presented in earlier sections, and in particular constraints on Γ and Δ from tracer ages, may be sensitive to our assumed functional form. For these cases an alternative

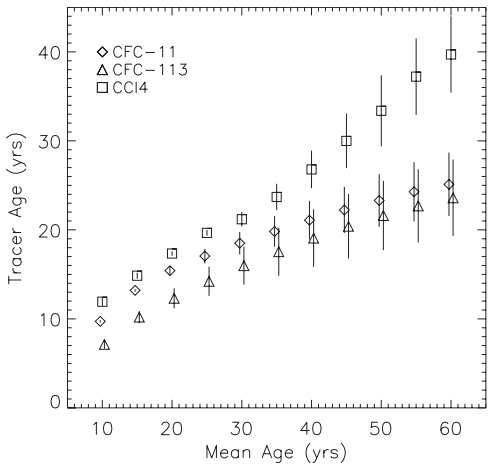


Figure 11. Variation of the mean and standard deviation of the distributions of halocarbon tracer ages for families of two-IG TTD with mean age for $\Gamma = 10-60$ years and $\Delta = 2\Gamma/3$. Symbols correspond to mean values and the vertical lines to ±1 standard deviation about this mean. When plotted, the mean ages for CFC-11 and CFC-113 have been increased and decreased, respectively, by 0.5 years for clarity.

approach is to split the TTD into two components, a young component sampled by the tracers and an old part that is tracer free [e.g., *Klatt et al.*, 2002; A. Putzka, unpublished manuscript, 1999].

8. Concluding Remarks

[76] Using the concept of a distribution of transit times we have performed a systematic examination of the relationship between the timescales (“ages”) derived from different tracers, for a wide range of flow conditions.

[77] From this analysis several important results can be drawn that need to considered, but are often not, when interpreting tracer ages.

[78] 1. Tracer ages are not, in general, fundamental timescales of the flow. Tracer ages depend on the time dependence of the surface boundary conditions and/or the timescale for decay as well as the distribution of transit times (which is a fundamental descriptor of the flow), and only in very special cases are the tracer ages equal to a timescale that has a simple relationship to the TTD and is independent of the tracer itself. One such case occurs when the tracer growth/decay is linear over the range of transit times, whence the tracer age approximates the mean transit time. This is true, for example, for measurements of CFC-11 and CFC-12 in the 1990s in water masses with young and narrow TTDs ($\Gamma < 20$ years and $\Delta < 10$ years).

[79] 2. Close agreement of ages from two tracers does not imply a lack of mixing or that the tracer age equals even approximately the mean age or the age of a third tracer. For example, while observations show close agreement between CFC-11 and tritium-helium ages [*Doney et al.*, 1997; *Frank et al.*, 1998], one cannot conclude that mixing is weak or that these tracer ages equal the mean age. Both τ_{CFC11} and τ_{He} can be approximately 15 years (as observed in the above studies) for TTD with $\Gamma \approx 30$ years and $\Delta \approx 30$ years, and for these TTD the time lag in anthropogenic carbon τ_{CO2} is around 20 years (see Figure 4). For older water masses the difference in ages can be even larger, for example, for $\tau_{CFC12} \approx 30$ years, and $\tau_{He} \approx 20$ years in 1988 (again as observed) Γ can be as large as 100 years and τ_{CO2} around 70 years.

[80] 3. Significant temporal variations in tracer ages can occur for stationary transport. For example, it is possible for a time-invariant TTD to produce temporal changes in CFC-11, CFC-12, and tritium-helium ages similar to those recently reported for the North Pacific and North Atlantic oceans [e.g., *Doney et al.*, 1998; *Robbins and Jenkins*, 1998; *Watanabe et al.*, 2001]. This means that the temporal changes in tracer ages do not necessarily imply temporal changes in the circulation.

[81] The systematic study of the age-age relationships also gives guidance on whether multiple tracers can be used to constrain the TTD. This analysis indicates that the degree to which Γ and Δ can be constrained depends on both the flow characteristics and the available tracers. For flows with little mixing tight constraints on Γ and Δ may be obtained (with $\Delta \approx 0$), whereas for flows with significant mixing a lower bound on Γ and Δ may be all that is possible. For young water masses (tracer ages less than 15 years) the most commonly measured tracers CFC-11, CFC-12, and tritium-helium provide very similar information on the TTD. However, CFC-113, CCl_4, and SF_6 provide different information (because of their different source histories), and measurements of one of these tracers with CFC-11 (or CFC-12) may constrain the TTD to a small region of (Γ, Δ) space. For older water masses CFC-11, CFC-12, and tritium-helium provide somewhat different information on the TTD, and the combination of CFC-11 (or CFC-12) ages with tritium-helium age may constrain (Γ, Δ) more than a single tracer age. Although CFC-113 and SF_6 ages cannot be defined for older water, CCl_4 can still provide a very useful constraint on the TTD.

[82] The preliminary analysis of actual tracer age measurements in section 6.2 suggests that in some situations available ocean and lake measurements of transient tracers provide enough information to constrain aspects of the TTD. For example, this analysis suggests that ocean water masses with CFC-11 ages greater than 20 years have broad TTD, with mean ages much larger than the tracer ages. We plan to examine this further using available tracer measurements (e.g., WOCE measurements). At this stage we have considered only transient tracer measurements and have not considered any other aspects of the flow (e.g., hydrographic or steady tracer measurements, or fluid dynamical models). Incorporation of such information into the analysis of age data may reduce the range of possible TTD that can produce given tracer ages.

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T. W. N. Haine and D. W. Waugh, Department of Earth and Planetary Sciences, Johns Hopkins University, Baltimore, MD 21218, USA. (thomas.haine@jhu.edu; waugh@jhu.edu)

T. M. Hall, NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA. (thall@giss.nasa.gov)